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Unified Formulation of X-Ray Diffraction and Refraction.

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(received 23 February 1993; accepted in final form 6 July 1993)

PACS. 61.10D – Theories of diffraction and scattering.

PACS. 41.10H – Electromagnetic waves: theory.

PACS. 68.90 – Other topics in the structure and nonelectronic properties of surfaces and thin films.

Abstract. – A novel formulation of X-ray diffraction and refraction is derived from the dynamical theory of X-ray diffraction. All the expressions of the dispersion relation, wave field amplitudes and reflectivities are given in analytic forms which are valid for the whole angular range of the symmetric diffraction geometry. In addition to putting Fresnel's reflectivity and Darwin's curve into a unified scheme, this framework provides a precise physical picture of diffraction from periodic media.

X-ray diffraction from crystals is a well-known phenomenon which has been utilized in many fields of research as a powerful tool for probing atomic structures. Along with the development of its applications, the diffraction theory also evolves from the geometric theory of Bragg and Laue diffraction to the kinematical theory and dynamical theory [1-7]. These theories were originally constructed for wide-angle X-ray diffraction and extended to the asymmetric Bragg case at a small incident angle [8-10]. Since the advent of synchrotron radiation, techniques of X-ray reflectivity measurement and grazing-incidence X-ray diffraction (GIXD) became useful in surface and interface studies [11,12], while the conventional theories have been modified to take care of in-plane diffraction [13,14] and surface-normal scan [15] at grazing incidence. However, due to approximations usually employed, there are practical situations that the current dynamical theory cannot fit well in describing diffraction phenomena. For example, i) in a general Bragg case, if the Bragg angle is close to the critical angle of total reflection, the conventional theory is incomplete since it is used to exclude the specular reflection [9,16]; ii) in surface normal scans of GIXD, the small-angle approximation breaks down, since the exit diffraction angle keeps no longer small [14].

In this letter, we present an exact geometrical construction of the dispersion surface and a smooth connection between optical refraction and X-ray diffraction. A set of analytic formulae of X-ray diffraction is derived from the plane-wave electromagnetic theory and the formulation is readily applied to the Bragg diffraction in general.

Considering the wave equation of X-rays in a crystal, wave fields are in the form of a

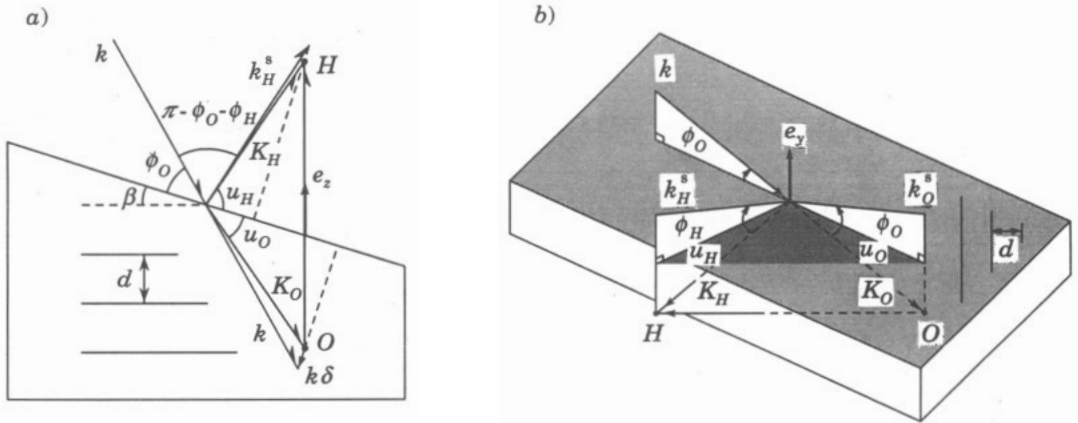


Fig. 1. - Geometry of a) asymmetric Bragg diffraction where $\overrightarrow{OH} \parallel \hat{e}_z$, and b) symmetric grazing-incidence X-ray diffraction where $\overrightarrow{OH} \perp \hat{e}_y$.

Bloch function, whereas the charge density is expanded by the Fourier components χ_G of the electric susceptibility. The fundamental equation of wave fields under the 2-beam approximation is [4, 5, 7]

$$(\chi_O - 2\varepsilon_O)E_O + C^P \chi_H E_H = 0, \quad C^P \chi_H E_O + (\chi_O - 2\varepsilon_H)E_H = 0, \quad (1)$$

where $\chi_G = -\Gamma F_G$, $-2\varepsilon_G = (k^2 - K_G^2)/k^2$, with $G = O$ standing for the incident direction while $G = H$ for the diffracted direction, and C^P is the polarization factor ($P = \sigma, \pi$): $C^\sigma = 1$ for the σ -polarized E_O^σ and E_H^σ , and $C^\pi \sim \cos 2\theta_B$ for the π -polarized E 's. We follow the convention: E_G^σ perpendicular to the plane of incidence. F_G is the structure factor and $\Gamma = r_e \lambda^2 / (\pi V)$: r_e is the classical electron radius, λ the incident-X-ray wavelength, and V the unit cell volume. θ_B is the geometric Bragg angle determined by the Bragg law: $2d \sin \theta_B = \lambda$, where d is the spacing of Bragg planes. As shown in fig. 1, \vec{k} is the wave vector in vacuum ($k = 1/\lambda$) and \vec{K}_G is the wave vector inside the crystal satisfying the Laue diffraction condition: $\vec{K}_H = \vec{K}_O + \overrightarrow{OH}$. The accommodation $k\delta$ between \vec{k} and \vec{K}_O is $\delta = -(\sin \phi_O - n_O \sin u_O)$, where ϕ_O is the incident angle, u_G is the angle between \vec{K}_G and the crystal surface, and $n_G \equiv K_G/k$.

We first consider a symmetric Bragg case where the crystal cut angle β is null, fig. 1a), where the exit ϕ_H is equal to ϕ_O . By introducing a proper variable for the resonance failure of diffraction: $\alpha_H = (K_H^2 - K_O^2)/k^2$, instead of α which is defined by $[(\vec{k} + \overrightarrow{OH})^2 - k^2]/k^2$, the null secular determinant of eq. (1) for non-trivial solutions is thus

$$(\chi_O + \sin^2 \phi_C)(\chi_O + \sin^2 \phi_C - \alpha_H) - C^P \chi_H C^P \chi_H = 0. \quad (2)$$

The characteristic angle ϕ_C is related to the generalized refractive index n_O by $n_O = \cos \phi_C$.

According to the identities: $\sin^2 \phi_C = -2\delta \sin \phi_O - \delta^2$ and $\alpha_H = \alpha - 4 \sin \theta_B \delta$, eq. (2) can be written as a 4th-order polynomial equation of δ [10]. Such an equation in principle can be analytically solved, however, the cumbersome expression of δ obscures the geometric feature of Bragg diffraction. In fact, since α_H is proportional to the coordinate variable:

$z = -\alpha_H/(4 \sin \theta_B)$, there is a better way to reformulate eq. (2) in terms of z ,

$$z^4 - 2[\sin^2 \theta_B + (\sin^2 \phi_O + \chi_O)]z^2 + [(\sin^2 \phi_O + \chi_O - \sin^2 \theta_B)^2 - C^P \chi_H C^P \chi_{\bar{H}}] = 0. \quad (3)$$

Two identities: $n_O \sin u_O = (\sin^2 \phi_O - \sin^2 \phi_C)^{1/2}$ and $\alpha_H = -4 \sin \theta_B (n_O \sin u_O - \sin \theta_B)$ have been used in deriving eq. (3). In short, with the geometric relations among the wave vectors k and K 's, z is equal to $n_O \sin u_O - \sin \theta_B$; both the diffraction and refraction are then taken into account in this variable z .

Due to the simple biquadratic feature, the dispersion relation of eq. (3) can be further decomposed into two hyperbola equations,

$$Y^2 - Z^2 = 1 \quad \text{and} \quad (Y + 2 \sin \theta_B/U)^2 - Z^2 = 1, \quad (4)$$

where $ZU = z$, $YU = (\sin^2 \phi_O + \chi_O + U^2)^{1/2} - \sin \theta_B$, and $U = (C^P \chi_H C^P \chi_{\bar{H}})^{1/2}/(2 \sin \theta_B)$. Equivalent to ϕ_O , we get the solution of eq. (3) as a function of the corrected Darwin's angular variable Y :

$$n_O \sin u_O = \sin \theta_B \mp \sqrt{Y^2 - 1} U \quad \text{or} \quad \sin \theta_B \mp \sqrt{(YU + 2 \sin \theta_B)^2 - U^2}. \quad (5)$$

For a given incident angle ϕ_O (*i.e.* Y), the real part of $n_O \sin u_O$ determines the position of tie points on the dispersion surface, while the imaginary part yields the absorption. To illustrate the difference between the present formulation and the conventional one without losing generality, for brevity, the imaginary parts of χ 's are neglected and the dispersion surface of each polarization can be analytically constructed as the dashed curve in fig. 2.

The dispersion surface consists of two deformed spherical surfaces centred at the

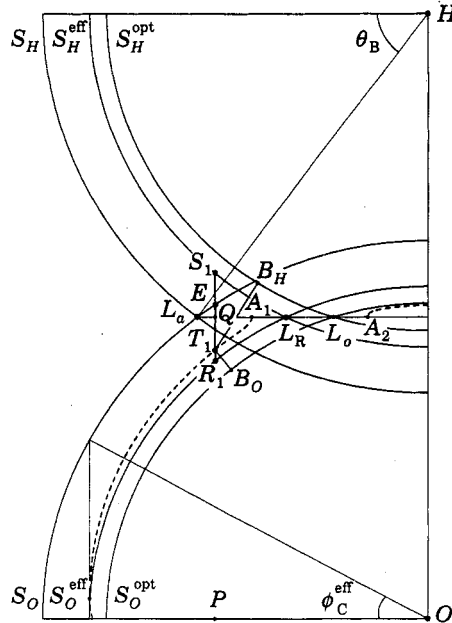


Fig. 2. - Schematic representation of the dispersion curve for the symmetric Bragg diffraction. The incident angle is determined by $\phi_O \equiv \angle EOP$. Since the surface normal is parallel to \overrightarrow{OH} , tie points T 's are then determined by the intersection of the dispersion curves with a line parallel to \overrightarrow{OH} through the entrance point E .

reciprocal lattice points O and H with the related radii K_O and K_H . Near the intersection of the two spherical surfaces, the curvature of the dispersion surface is modified by diffraction effects. Figure 2 shows the details of this intersection region cut by the plane of incidence, where the scale near the intersection region has been exaggerated. L_a and L_o are the Laue point and the Lorentz point where $\overline{L_a G} = k$ and $\overline{L_o G} = k[(1 + \chi_O)^{1/2}]$. L_R is the point on $\overline{L_a L_o}$ which is very close to L_o , since $\overline{L_R G} = k[(1 + \chi_O + U^2)^{1/2}]$. Three pairs of concentric circles, $\{S_O, S_H\}$, $\{S_O^{\text{opt}}, S_H^{\text{opt}}\}$, and $\{S_O^{\text{eff}}, S_H^{\text{eff}}\}$, are «asymptotic» curves of the dashed dispersion surface, but S_G^{eff} intersects the dashed curve, since \overline{EP} is a tangent of both the dashed curve and S_O^{eff} at the angle $\phi_O = \phi_C^{\text{eff}} = \arcsin[(-\chi_O - U^2)^{1/2}]$ (i.e. the effective critical angle).

For a given incident ϕ_O , a line passing through the points S_1, E, Q, T_1, R_1 and normal to the crystal surface (as shown in fig. 2) determines at most four tie points T 's on the dispersion curve according to the four solutions of eq. (5). However, for an infinitely thick crystal, only two points (T_1 and T_2) with Poynting vectors toward the interior of the crystal satisfy the law of energy conservation. As ϕ_O increases, the excited T_i ($i = 1$ to 4) moves along the dashed curve following the dispersion relation

$$\overline{T_i R_i} \cdot \overline{T_i S_i} = k^2 C^P \chi_H C^P \chi_{\bar{H}} / (2 \sin \theta_B)^2. \quad (6)$$

This relation is valid for all the angular range except in the discontinuous region between A_1 and A_2 , which corresponds to Darwin's plateau of total reflection. Recall that the conventional result: $\overline{T_1 B_O} \cdot \overline{T_1 B_H} \approx k^2 C^P \chi_H C^P \chi_{\bar{H}} / 4$ ($\overline{T_1 B_G}$ normal to S_G^{opt}) is valid only near the region of Darwin's diffraction plateau.

The dashed dispersion curve asymptotically approaches the effective circle S_O^{eff} at the end as T_1 below A_1 , while it comes close to the optical one S_O^{opt} at the other end as T_1 above A_2 . Because the radius of S_G^{eff} is $k[(1 + \chi_O + U^2)^{1/2}]$, the diffraction comes into play in effecting the refraction due to the presence of U , where the critical angle of total reflection changes to ϕ_C^{eff} , but only with 10^{-5} correction in magnitude. Nevertheless, U is the essential unit ratio in realizing the geometry of dispersion relation, eqs. (4) and (6). Not only the resonance failure α_H has its own geometric meaning (i.e. $ZU\hat{e}_z = \overrightarrow{QT_1}/k$), but also the corrected Darwin's variable: $YU\hat{e}_z = \overrightarrow{QR_1}/k$.

Geometric differences between conventional theories and the present formulation are listed in table I. The resonance angle ϕ_R is the entrance angular position corresponding to the

TABLE I. - Comparison between conventional theories and the present formulation for symmetric Bragg diffraction.

	Conventional theories	Present formulation
resonance failure	$\alpha \approx -2(\phi_O - \theta_B) \sin 2\theta_B$	$\alpha_H = -4 \sin \theta_B (n_O \sin u_O - \sin \theta_B) \rightarrow$ $\rightarrow -2(\phi_O - \theta_B) \sin 2\theta_B$
Darwin's variable	$Y \equiv (\chi_O - \alpha/2) / C^P \chi_H $	$Y \equiv (\sqrt{\sin^2 \phi_O + \chi_O + U^2} - \sin \theta_B) / U \rightarrow$ $\rightarrow (\chi_O - \alpha/2 + U^2) / C^P \chi_H $
dispersion relation	$\overline{T_1 B_O} \cdot \overline{T_1 B_H} \approx k^2 C^P \chi_H C^P \chi_{\bar{H}} / 4$	$\overline{T_1 R_1} \cdot \overline{T_1 S_1} = k^2 C^P \chi_H C^P \chi_{\bar{H}} / (2 \sin \theta_B)^2$
critical angle	$\sin \phi_C^{\text{opt}} = \sqrt{-\chi_O}$	$\sin \phi_C^{\text{eff}} = \sqrt{-\chi_O - U^2}$
resonance angle (Bragg angle shifting)	$\Delta\theta_{RB} \equiv \phi_R - \theta_B \approx -\chi_O / \sin^2 \theta_B$	$\sin^2 \phi_R = \sin^2 \theta_B + \sin^2 \phi_C^{\text{eff}} \Rightarrow$ $\Rightarrow \Delta\theta_{RB} \rightarrow (-\chi_O - U^2) / \sin 2\theta_B$

centre ($Y = 0$) of Darwin's plateau, where a surface normal passes through the entrance point E and resonant point L_R . An exact expression of the Bragg angle shifting is then obtained:

$$\sin^2 \phi_R = \sin^2 \theta_B + \sin^2 \phi_C^{\text{eff}}. \quad (7)$$

It reduces to Darwin's result [2]: $\Delta\theta_{RB} \equiv \phi_R - \theta_B \approx -\chi_O/\sin 2\theta_B$, since ϕ_C^{eff} is small. Since the dispersion surface is independent of the cut angle β , the same picture can be extended to the asymmetric Bragg case. A close-form expression is derived:

$$\sin^2 \phi_R = \sin^2 \phi_B + \cos^2 \beta \sin^2 \phi_C^{\text{eff}} + \sin 2\beta \sin \theta_B (\cos \theta_B - \cos \theta_R), \quad (8)$$

where $\phi_B \equiv \theta_B - \beta$ and θ_R is ϕ_R at $\beta = 0$. Because the difference between ϕ_R and ϕ_B is small, eq. (8) reduces to Rustichelli's form [9]

$$\Delta\phi_{RB} \equiv \phi_R - \phi_B \approx \left\{ -\sin \phi_B + \left[\sin^2 \phi_B + \frac{\cos \beta}{\cos \theta_B} \sin^2 \phi_C^{\text{eff}} \cos \phi_B \right]^{1/2} \right\} / \cos \phi_B, \quad (9)$$

which has been well confirmed in a full numerical calculation [10].

The ratios between wave field amplitudes are derived from eq. (1) as $v(i) = E_H(i)/E_O(i) = [Z \pm (Z^2 + 1)^{1/2}]/[(C^P \chi_H C^P \chi_H)^{1/2}/(C^P \chi_H)]$ for two modes: $i = 1, 2$. The absolute wave field amplitudes are determined by the boundary condition for the continuities of the normal components of the electric displacements and the tangential components of the magnetic fields at the crystal surface:

$$\sum_{i=1}^2 E_G(i) = E(G) + E_G^s, \quad \sum_{i=1}^2 n_G(i) \sin u_G(i) E_G(i) = \sin \phi_G (E(G) - E_G^s) \quad (10)$$

for the σ -polarized waves. $E(O) = E_{\text{inc}}$ and $E(H) = 0$. E_{inc} is the incident-wave field amplitude and E_G^s is the surface-reflected one. Equation (10) leads to

$$E_O(i) = G(i) \{ 2 \sin \phi_O / [\sin \phi_O + n_O(i) \sin u_O(i)] \} E_{\text{inc}}, \quad (11)$$

where $G(1) = 1/(1+t)$ and $G(2) = t/(1+t)$ are geometric weighting factors with $t = -[v(1)w_O(2)w_H(1)]/[v(2)w_O(1)w_H(2)]$ and $w_G(i) = \sin \phi_G + n_G(i) \sin u_G(i)$. The mirror-reflected E_O^s and Bragg-diffracted E_H^s are derived as

$$E_O^s = \sum_{i=1}^2 G(i) \{ [\sin \phi_O - n_O(i) \sin u_O(i)] / [\sin \phi_O + n_O(i) \sin u_O(i)] \} E_{\text{inc}}, \quad (12)$$

$$E_H^s = \sum_{i=1}^2 v(i) E_O(i). \quad (13)$$

The reflected powers of the mirror reflection and Bragg diffraction are then

$$P_O |E_O^s/E_{\text{inc}}|^2 \quad \text{and} \quad P_H = \text{Re} \{ (\sin \phi_H / \sin \phi_O) \} |E_H^s/E_{\text{inc}}|^2. \quad (14)$$

The terms in the braces of eqs. (11) and (12) are Fresnel's transmission and reflection coefficients, and the sum of $G(1)$ and $G(2)$ is unity. It is worth noting that the mode excitation is merely a product of the geometric factor and Fresnel's coefficient. Similar results for the π -polarized waves can be obtained in a straightforward manner.

The same derivation can be applied to the case of Laue diffraction geometry. Instead of dealing with the Laue transmission, we consider a more general case involving GIXD as shown in fig. 1b), where OH lies in the plane parallel to the crystal surface. The same equations (10)-(14) can be obtained from the same procedure which cover the results derived

from the small-angle approximation, i.e. $\sin \phi_G$ replaced by ϕ_G . As a comparison,

$$P_H(\phi_O) = \left| \phi_O \left[\frac{\sqrt{\phi_O^2 + \chi_O + \chi_H} - \sqrt{\phi_O^2 + \chi_O - \chi_H}}{(\phi_O + \sqrt{\phi_O^2 + \chi_O + \chi_H})(\phi_O + \sqrt{\phi_O^2 + \chi_O - \chi_H})} \right] \right|^2, \quad (15)$$

derived by Afanas'ev and Melkonyan [14] for « σ »-polarized waves under $\alpha_H = 0$, while a more informative form is read from our approach,

$$P_H^{\tau}(\phi_O) = \left| \frac{1}{2} \left\{ \frac{2 \sin \phi_O}{\sin \phi_O + \sqrt{\sin^2 \phi_O + \chi_O - |C^{\tau} \chi_H|}} \right\} - \frac{1}{2} \left\{ \frac{2 \sin \phi_O}{\sin \phi_O + \sqrt{\sin^2 \phi_O + \chi_O + |C^{\tau} \chi_H|}} \right\} \right|^2. \quad (16)$$

Explicitly, at the exact resonant condition, an interference between the two equally weighted refraction modes gives the specular Bragg diffraction.

In conclusion, a unified formulation of X-ray diffraction and refraction has been presented in the symmetric cases. Both Darwin's diffraction curve and Fresnel's optical reflectivity are taken into account in a standard electromagnetic theory. By contrast with the conventional theories which are only valid in a narrow region around Darwin's plateau, our present work provides a rigorous treatment for the dynamical theory of diffraction and gives a precise physical picture of diffraction from periodic media.

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The authors are much indebted to the National Science Council of the Republic of China for financial support. HHH would like to show his high appreciation to KENG S. LIANG for initially drawing our attention to GIXD.

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